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Atmospheric Metal Pollution Monitored by Spherical Moss Bags: A Case Study of Armadale

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To supplement epidemiological investigations into the mortality from respiratory cancer in the small industrial town of Armadale, central Scotland, spherical moss bags were used to study the deposition of atmospheric metal pollution there during a period of 17 months. High concentrations of most metals were found in areas close to the local steel foundry. High concentrations of some metals were also found in the north/northeast of the town. Temporal variations in the metal deposition patterns during the survey-period were observed. By means of statistical analyses, those metals were indicated which were probably emitted from the steel foundry. The scientific and financial advantages of using this method of low technology sampling in epidemiological studies are discussed.

Introduction

Armadale is a long-established industrial town with a population of about 8000 and an area of less than 3 km^2. It is situated on exposed moorland in central Scotland, midway between Edinburgh and Glasgow, and about 20 km from each. A foundry, situated in the southeast quadrant of the town, produces special steels. Epidemiological studies have revealed an association between the mortality from lung cancer and atmospheric metal pollution (*1–3*). To investigate this association further, more detailed systematic investigations of air pollution by metals were required. Because the cost of conventional equipment was too high to allow air pollution to be sampled at the many sites required, it was decided to use less expensive low technology samplers.

Of the low technology samplers, mosses and lichens have been shown to have the most desirable properties (*4–7*). During the last decade, a new type of transplanted biomonitor, the moss bag, has been developed which overcomes many of the limitations of live indigenous samplers (*8–10*) while retaining many of their qualities which are desirable for sampling. In the moss bag, the moss has usually been removed from its natural habitat, killed, cleaned, leached of its existing exchangeable metal content, and finally suspended in a nylon net container in the field.

Moss bags can be spherical, horizontal and flat, or vertical and flat. The shape of the spherical moss bags (SMB) allows them to collect metal particles relatively equally from all directions, and metal concentrations in them are higher and more consistent than those in flat moss bags (*11*). Many species of moss, including *Hypnum cupressiforme*, have been used for the construction of moss bags (*12*); however, Sphagnum moss has been used most frequently and has many advantages.

The main advantages of Sphagnum moss as a monitor of airborne metals are as follows. (1) This moss has a greater proportion, by weight, of cation exchange sites than any other recorded species of plants (*13*). The leaves of this moss have a relatively low protoplasmic content, and hence more of its structure is available as cation exchange sites. (2) Two-thirds of the dry weight of Sphagnum is composed of leaves (*13*); the ratio (dry weight) of leaves to branches is 6:1, which provides a very large surface area on which to entrap and retain metal-rich particles. (3) The leaves of most species are arranged spirally around the stem, which enhances the capture of particles from the ambient atmosphere. (4) The dead hyaline cells, of which this moss is mostly composed, are covered in surface pores which may facilitate the physical retention of particles. (5) With the leaves being only one cell thick, a very large proportion of the structure is in contact with the atmosphere. (6) Because Sphagnum can receive all of its nutrients from the atmosphere, it has developed very efficient mechanisms for accumulating metals over the entire exposed surface; for example, being ectohydric, it can absorb cations from very dilute solutions such as rainwater (*13*). (7) This moss can hold a water volume which exceeds its structural weight by more than 20-fold, facilitating ion retention (*14*). (8) Its natural habitat is usually acidic, which enhances ion exchange even without acid-washing. (9) It can have 20–30% of its dry weight com-

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posed of uronic acids; these acids release H^+ ions, ensuring both a low pH and empty exchange sites. (*10*) The relatively large amount of information published about the behavior and characteristics of this moss can be applied to sampling surveys (*8*).

The transplanted mosses can be acid-washed, water-washed, or left untreated. Acid-washed mosses give the highest and most consistent metal concentrations (*11*), probably because they are not affected by varying metabolic activities, because they have the most consistent background metal concentrations, and because they also have the highest number of free cation exchange sites. In most investigations in which moss bags have been employed, the acid-washed type of Sphagnum moss was used (*15–17*).

For the present survey, therefore, SMB containing acid-washed Sphagnum moss were chosen as samplers.

Methods

Sphagnum palustre was collected from a remote moorland to ensure that the moss had not been damaged by air pollution; with only low concentrations of metals in the moss, only small amounts of acid are needed to leach out the exchangeable metals.

At the laboratory, the moss was rigorously cleaned to remove extraneous material. Only green shoots of moss were selected, thereby excluding the decaying shoots in which cation exchange sites had been at least partly destroyed. Particles of detritus and soil were removed by rinsing the moss several times under a jet of deionized water. The moss was submerged in a plastic bin which had been deleaded and filled with 1 N HNO_3 (analytical reagent grade); after remaining in this solution for 3 days, the moss was rinsed three times under a jet of deionized water. After the excess water had been squeezed out, the moist moss was stored in a plastic bag.

The containers for the SMB were made from a nylon net, a 2 mm mesh size, cut in circles of 15 cm diameter; this size of nylon circle allowed the moss to be loosely packed and thus gave good access to air pollutants (*18*). Approximately 0.15 g (dry weight) of prepared moss was placed in the center of the nylon circle; the circle was folded in half and the edges were joined by nylon thread. The thread was then drawn together to form a sphere of diameter approximately 2.5 cm (*18*). This net bag containing the moss was itself enclosed in a nylon hair-bun net, to give the SMB sufficient strength to withstand adverse weather conditions.

The survey, which started in May 1981, lasted 17 months. Each SMB was attached by means of plastic string glued to a sampling head; the head was fixed to a bamboo cane, 2 m above ground level. Before exposure the SMB were sprayed with deionized water to maximize their capacity for collecting metal ions. The SMB were exposed at 47 sites in Armadale (*19*) for each of eight batches (exposure periods); seven of the batches lasted for 2 months each, batch 4 lasting for 3 months because bad weather prevented fieldwork.

After exposure, the SMB were cut from the sampling heads and were carefully transported to the laboratory in plastic bags. In the laboratory, the moss was removed from the nylon net containers and oven-dried at 80°C to constant weight. The mosses were wet-digested, and the concentrations of nine metals were analyzed by using atomic absorption spectrophotometry; technical details have been described elsewhere (*10,20*).

The background metal concentrations, which varied within 10% between the different loads of the unexposed moss, were subtracted from the concentrations in the exposed moss.

For each metal and site, the concentrations in each batch and the mean concentrations for the eight batches combined were calculated; the mean concentrations were displayed cartographically. Grid maps (Fig. 1) were used to show the observed values at the individual sampling sites; contour maps, produced by means of the statistical package GLIM, illustrated pollution flow by estimating relative concentrations for all areas of the town (Fig. 2). Furthermore, in order to quantify the relationship between the concentrations of metals at the various sites and the distance of the sites from the foundry (irrespective of orientation), the area in and around Armadale was divided into six squares of different sizes, each centered on the foundry (Fig. 3); the mean concentrations of metals at sites within the peripheral areas of each square were calculated for the entire survey and for the individual batches. Examples of the use of these map patterns in other surveys have been given elsewhere (*9,10,19,20*).

Analysis of variance (ANOVA) was used to discover if the mean concentrations of each metal differed significantly between the batches, and also between the six areas of the square map pattern; ANOVA was also used to test the concentration gradient in the square map pattern for linearity. In order to indicate those metals sharing a common source or sources, Spearman's rank correlation coefficients were generated between the concentrations of all metals over 47 sites for individual batches; these coefficients were also calculated for the concentrations of the same metal over 47 sites between the individual batches, in order to observe any fluctuations in the metal concentrations which could have resulted from meteorological variations or from alterations in the output from that metal's source.

The approximate cost of the materials used in the preparation and analysis of the SMB was noted.

Results

General Statistical Analyses

Iron, Mn, Zn, Pb, and Cu were found at most of the 47 sites in all batches; Cr was not present at any site in batches 6 and 7, nor was Ni in batches 5 to 8; Cd and Co were never accumulated.

The mean concentrations of each metal for the whole town varied between batches (Table 1). In all batches,

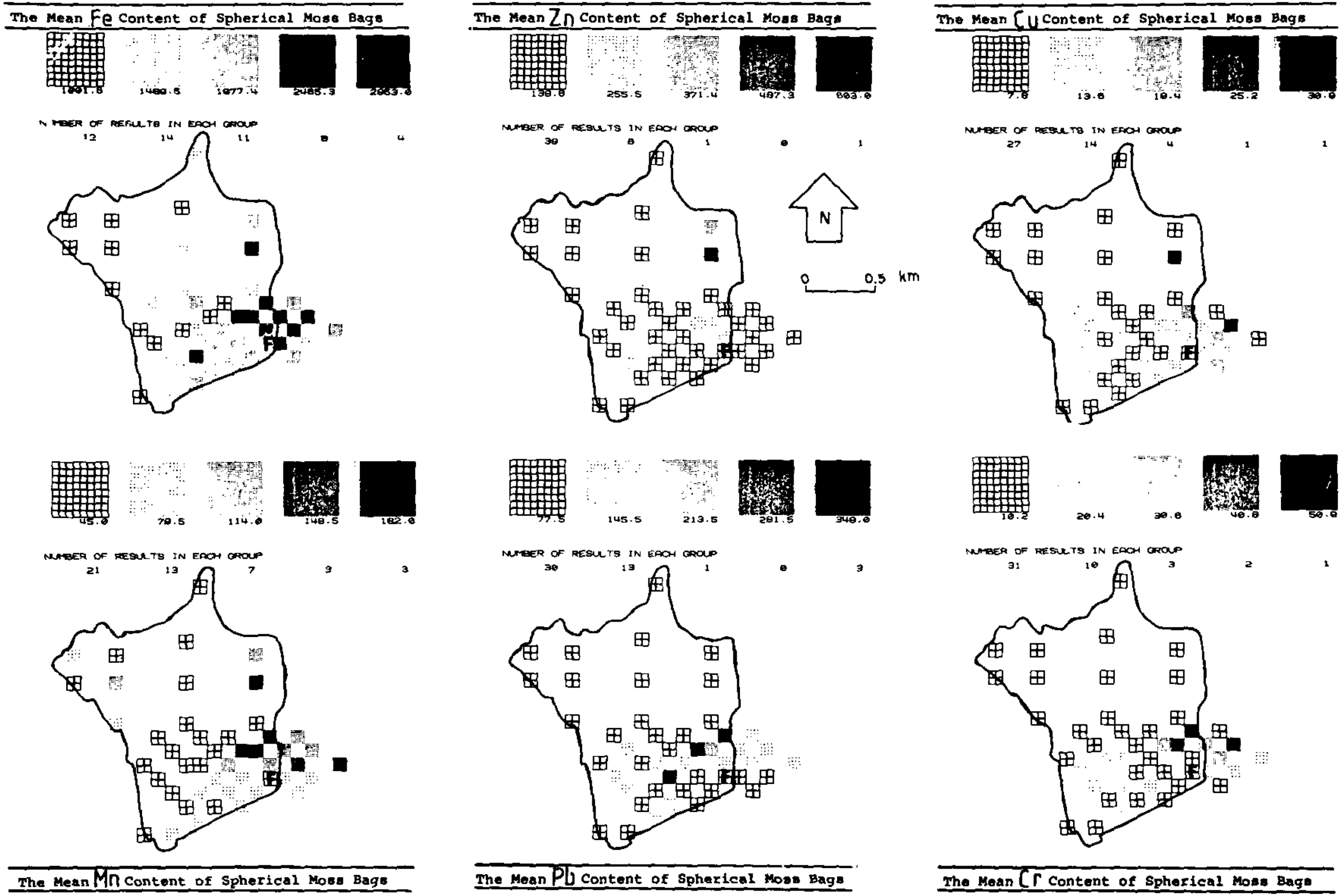

FIGURE 1. Grid maps showing the mean concentrations of each metal except Ni over all batches, collected in spherical moss bags; the site of the foundry lies directly to the north of "F".

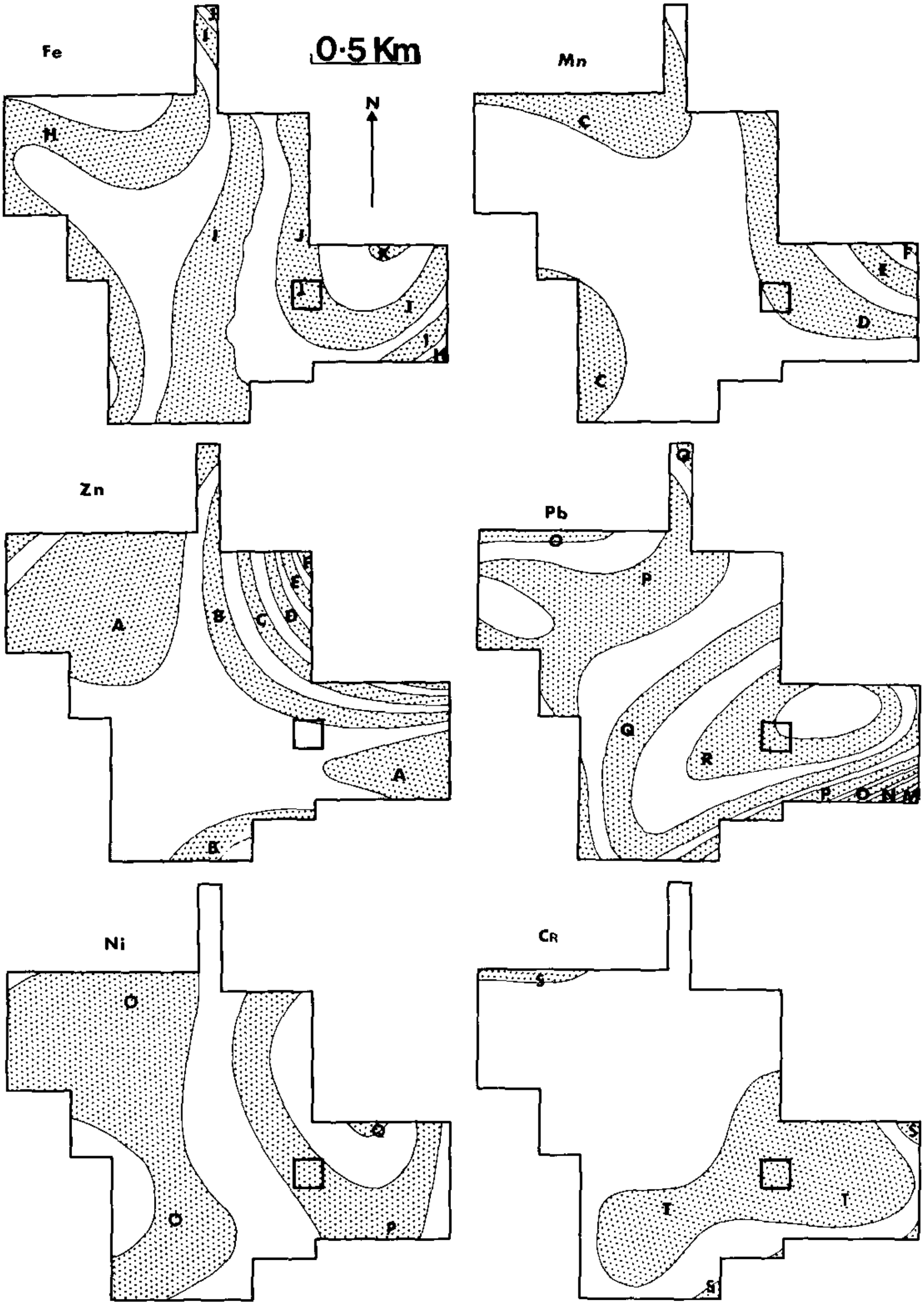

FIGURE 2. Maps showing the flow of pollution by Fe, Mn, Zn, Pb, Cr, and Ni. The relative estimated concentrations are represented by letters, the higher relative concentrations being denoted by the letters lower in the alphabet; the square represents the approximate site of the foundry.

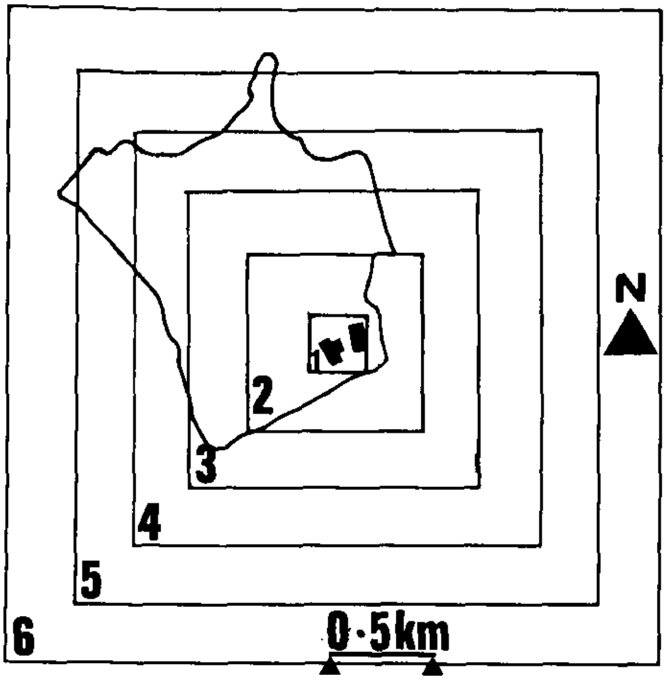

FIGURE 3. The square map pattern; the foundry buildings are in the central square.

the mean, maximum and minimum concentrations of Fe exceeded those parameters of the other metals, the values of Mn, Zn, and Pb concentrations were notably lower, while the values of Cu, Cr and Ni were lowest.

Of the coefficients of variation used to compare the variability of the concentrations of the different metals, those of Fe, which had the highest concentrations, were only moderate; those of Ni, Cu, Pb, Zn, and Mn were higher; and those of Cr, which was found in low concentrations, most frequently were the highest. The skewness values, used to select the most appropriate correlation technique, indicated that most of the metal concentrations in the eight batches were not normally distributed among the 47 sites.

The two-way ANOVA revealed that, for each metal except Ni, the mean concentrations for the whole town differed highly significantly ($p < 0.005$) between the batches, and that there were highly significant differences between the mean concentrations of the metals during the whole survey at individual sites.

Mapping Patterns

Grid Maps. On the maps showing the mean concentrations of each metal (except Ni) during the whole survey (Fig. 1), the highest concentrations were mainly close to the foundry; high mean concentrations were also found in the town's northeast section; for Zn, the site with the highest concentration was in Armadale's northeast section, with slightly elevated values along a west–east axis close to the foundry.

In the individual batches, different metals showed different concentration patterns; the patterns of a given metal varied between batches. Nevertheless, again many maps showed moderate to high concentrations of metals in the vicinity of the foundry. Many of the lowest concentrations were at the town's periphery, with the exception of the southeast periphery near the foundry.

Estimated Concentration Maps. Generally, the bands with the highest concentrations of metals were in the southeast quadrant of the town (Fig. 2). The pollution flow was seen to extend from the foundry to the east, towards the north of the town, and occasionally to the west of the foundry. (For Cu, only one band of estimated concentrations was demonstrated, indicating a narrow range of estimated concentrations throughout the town.)

Square Maps. Relatively high concentrations were most frequently present in the areas close to the foundry (Fig. 3) both in the whole survey (Table 2) and in the individual batches (Table 3). The concentrations usually declined discontinuously outwards from the foundry, Fe and Mn having the most pronounced and consistent gradients in the individual batches. An "umbrella effect" was seen for most metals in many batches, with the area closest to the foundry having concentrations lower than those in the adjacent area which was slightly more distant from the foundry. For Pb, most of the batches showed no concentration gradient.

The ANOVA showed that the mean concentrations of all metals except Pb differed highly significantly ($p < 0.005$) between the six areas; for Pb, the mean concentrations were only significantly different ($p < 0.05$). The analysis also revealed that the mean concentrations of each metal in the six consecutive areas were highly significantly linear ($p < 0.005$).

Correlations

For each batch, the intercorrelations between the concentrations of each metal calculated over 47 sites were mostly positive, showing that the metals had probably been emitted by a common source.

The percentages of metal pairs which were significantly correlated in all batches ranged from 30% to 93% (Table 4), the number of significant correlations being partly dependent upon the number of metals collected. The metal pairs that were significantly correlated most frequently were Mn/Fe, Mn/Zn, and Fe/Zn. Other metal pairs that were also significantly correlated for at least 50% of the batches were: Fe/Cu for six batches; Cu/Cr, Fe/Cr, Mn/Cu, and Mn/Pb for five batches each; and Mn/Cr and Fe/Pb for four batches each; Ni was significantly correlated with most metals for at least 50% of the batches in which it was collected.

The numbers of other metals with which a given metal was significantly correlated in individual batches varied between the metals within each batch and between the batches for the same metal (Table 5). For example, in batch 5 the numbers of significant correlations were high for all metals except Ni, whereas in batches 6 and 7 there were very few significant correlations. Iron and Mn were significantly correlated with the highest num-

Table 1. Spherical moss bags: the statistics of each metal and batch for the whole town.[a]

Metal	Batch	Number	Mean, μg/g	SD	CV, %	Min, μg/g	Max, μg/g	Skewness
Fe	1	40	541.8	293.6	54.2	103.2	1202.8	0.7
	2	37	334.4	292.8	87.6	78.3	1303.7	2.1
	3	43	1083.7	851.4	78.6	120.1	3705.6	1.5
	4	44	1960.1	1159.3	59.1	188.5	5859.2	1.0
	5	46	2047.5	1209.4	59.1	444.5	6297.7	1.8
	6	41	1710.6	1158.6	67.7	119.6	4987.5	0.9
	7	39	1827.6	1267.4	69.3	582.5	6463.5	2.0
	8	38	1783.4	784.5	44.0	737.5	3842.5	1.0
Mn	1	40	19.0	20.5	107.9	0.0	66.6	1.0
	2	37	15.2	30.6	201.3	0.0	180.8	4.8
	3	43	34.1	33.8	99.1	2.6	156.8	1.9
	4	3	103.7	129.1	124.5	6.5	813.2	4.3
	5	46	115.5	122.4	106.0	4.4	638.1	2.5
	6	1	122.0	99.4	81.5	0.0	354.5	0.9
	7	39	32.1	51.0	158.9	0.0	180.0	1.9
	8	38	30.2	30.1	99.7	0.0	140.5	1.8
Zn	1	40	23.3	29.0	124.5	0.0	131.0	2.1
	2	37	12.3	18.2	148.0	0.0	85.5	2.3
	3	43	58.8	30.4	51.7	12.5	154.9	1.3
	4	44	69.4	61.5	88.6	9.9	352.3	2.7
	5	46	114.0	251.6	220.7	0.0	1209.8	3.9
	6	41	344.6	120.6	35.0	0.0	592.1	−0.3
	7	39	50.3	72.3	143.7	0.0	355.0	2.5
	8	38	78.4	140.2	178.8	12.0	652.0	3.9
Pb	1	40	9.6	9.5	99.0	0.0	29.8	0.7
	2	37	8.5	5.3	62.4	0.0	24.5	1.0
	3	43	38.7	32.6	84.2	0.0	163.4	1.9
	4	44	48.8	42.5	87.1	0.0	149.2	1.1
	5	46	300.6	455.4	151.5	0.0	2042.5	2.4
	6	41	101.2	40.6	40.1	45.0	225.0	1.0
	7	39	17.8	69.9	392.7	0.0	430.5	5.7
	8	38	23.7	20.2	85.2	0.0	75.0	1.0
Cu	1	40	0.9	1.2	133.3	0.0	3.6	1.2
	2	37	7.9	9.3	117.7	0.0	45.4	2.6
	3	43	17.1	24.6	143.9	0.4	159.8	4.9
	4	44	2.6	3.6	138.5	0.0	16.3	2.2
	5	46	7.1	5.5	77.5	0.0	31.3	2.3
	6	41	22.6	17.4	77.0	0.0	68.0	0.7
	7	39	4.0	8.3	207.5	0.0	26.0	1.9
	8	38	3.6	5.7	158.3	0.0	23.0	2.4
Cr	1	40	3.0	6.0	200.0	0.0	27.1	3.1
	2	37	8.6	27.4	318.6	0.0	167.7	5.7
	3	43	5.9	9.7	164.4	0.0	40.0	2.1
	4	44	17.9	30.5	170.4	0.0	145.1	2.4
	5	46	10.7	18.6	173.8	0.0	86.0	2.8
Cr was not collected in batches 6 and 7								
	8	38	12.4	23.2	187.1	0.0	83.0	1.8
Ni[b]	1	40	1.5	1.4	93.3	0.0	5.3	0.7
	2	37	5.4	11.5	213.3	0.0	64.8	4.3
	3	43	7.1	8.9	125.4	0.0	33.5	1.3
	4	44	4.8	7.8	162.5	0.0	31.3	1.7

[a] Batch refers to the period of exposure; number refers to the number of moss bags collected for each batch; mean refers to the mean concentrations of the metals; SD refers to the standard deviation; CV refers to the coefficients of variation; and min and max refer to the *minimum and maximum concentrations found at any site during a particular batch.*

[b] Ni was not collected in batches 5 and 8.

ber of metals most frequently; Pb was often insignificantly correlated with most metals.

All of the Spearman's rank correlation coefficients between the mean metal concentrations in the whole town during individual batches were positive (Table 6); 20 of the 21 possible correlations were significant, 17 at $p < 0.005$ and 3 at $p < 0.05$; Cr/Zn, Ni/Zn, and Cu/Mn were within the latter significance level. Only Cu/Zn was not significantly correlated.

In general, for only Fe and Mn were the concentra-

Table 2. Spherical moss bags: the mean concentrations of each metal over all batches in the six areas of the square map pattern.

	Metal concn, μg/g						
Area	Fe	Mn	Zn	Pb	Cu	Cr	Ni
1	1846.0	69.0	108.6	67.2	8.5	6.9	7.9
2	1776.0	80.8	93.5	120.5	9.5	15.6	6.9
3	1460.0	56.5	94.7	61.8	9.1	11.7	4.2
4	1118.0	41.9	127.6	50.9	7.6	3.7	3.0
5	692.9	45.8	61.0	26.1	5.0	0.7	1.7
6	950.6	46.9	58.7	28.3	5.2	1.0	1.4

tions in a given batch significantly correlated with the concentrations in the other batches (Table 7).

The total cost of the materials used to construct and analyze the SMBs during the whole survey was less than £100.

Discussion

By using SMB to sample pollution at 47 sites in Armadale, and by illustrating the results on grid maps, this study of atmospheric metal pollution revealed two main areas of metal deposition in the town, thereby confirming the results of work using other types of low technology sampler (*10,19*). These areas approximated to the two areas where clustering of lung cancer had been noted earlier (*3*); one area was in the vicinity of the steel foundry, but in the second area in the north-northeast (NNE) of Armadale local sources of metals could not be found. However, the patterns of northward flow of pollution illustrated by the estimated concentration maps indicated that the latter area could also have been subjected to air pollution from the foundry. This view was supported by a wind tunnel experiment which demonstrated the effect of the town's topography in promoting the northerly flow of that pollution from the foundry (*21*).

Within the individual batches, high concentrations of most metals were also close to the foundry, with an "umbrella effect" often apparent; moderate concentrations of Pb were near the main traffic route, low concentrations of metals at the town's periphery suggested that external sources of metal pollution were unimportant.

The use of the square map pattern indicated gradients of pollution by Fe, Mn, Cu, Cr, and Ni, and to a lesser extent Zn and Pb, with high values near the town's steel foundry; for several metals, the "umbrella effect" around the foundry was also quantitatively demonstrated.

That most metals had a common main source was further supported by the intercorrelations found between the concentrations for each metal in the whole town during the individual batches. Iron and Mn were significantly correlated with several metals in most batches; this had been expected because Fe and Mn are the accepted metal markers of the steel industry (*22*). The intercorrelations between the concentrations of a given metal for individual batches in the whole town was in agreement with the proposition that Fe, and to a lesser extent Mn, had a constant and stable source. The relatively few significant correlations of the remaining metals probably reflected their relatively low concentrations (for Cu, Cr, and Ni) and the variability of their source or sources (for Pb and Zn).

There were several possible reasons, other than proximity to sources of air pollution, for the variations in metal deposition patterns within each of the eight batches, and also between the batches for the same metal.

Meteorological factors caused some of the variability in the metal deposition patterns. Because the area of Armadale is only 2.6 km^2, variations in meteorological parameters other than wind flow were unlikely to have caused the differences in the metal concentrations throughout Armadale within any one batch. By contrast, some of the notable differences between the different batches undoubtedly resulted from changing meteorological conditions and, in particular, wind characteristics and precipitation. However, because of the complexity of the effects of meteorological parameters on the collection of metals in the SMB it is difficult to assess precisely how the concentrations were affected. For instance, higher wind speeds dilute the concentrations of metal particles in the air brought to an SMB; but they also cause a greater volume of polluted air to be filtered through the SMB, thereby favoring an increase in the concentrations of metals collected. In batches 4, 5, and 6, which were during the winter and spring, higher values of most metals were found. But how far that finding was influenced by the weather, rather than by changes in output, is not clear (unpublished observations).

Changes in the output of the metals from one or more sources during the eight batches must have caused some of the differences of the patterns between batches. However, there was no obvious relationship between the amounts of metals smelted by the foundry and the concentrations of metals collected in the individual batches (unpublished observations). The other less important sources of metal pollution within Armadale, such as brick production, petrol combustion, domestic coal combustion, and farming activities, were unlikely to have varied enough during the survey to account for the variations in the metal deposition patterns.

A small amount of the variation could have resulted from the inherent properties of the SMB (*11*). Firstly, because most of the metals are collected by cation exchange, there could be some competition for uptake between the metals; for example, Mn has a very low placing in the sequence of preferential uptake (*23*); however, it is unlikely that the concentrations of metals collected were high enough for this factor to have been significant. Secondly, although the moisture content of the SMB within the one batch was likely to be relatively uniform, that between the batches was likely to alter because of differing weather conditions, and these alterations could have affected the amounts of metals accumulated. Thirdly, several collections of Sphagnum

Table 3. Spherical moss bags: the metal concentration of each area and batch for the square map pattern.

Batch area	Metal concn, µg/g						
	Fe	Mn	Zn	Pb	Cu	Cr	Ni
B1							
1	658.2	31.1	16.3	3.1	0.3	1.9	1.9
2	727.8	34.4	37.3	8.8	1.1	2.8	2.1
3	547.1	14.9	21.3	9.5	1.0	4.7	1.1
4	401.0	6.8	16.7	14.6	0.9	2.9	0.9
5	243.5	2.5	5.5	8.0	0.3	0.3	1.8
6	308.2	9.4	19.3	10.9	2.0	1.3	1.3
B2							
1	596.9	23.6	42.3	7.7	18.9	3.1	10.5
2	511.4	30.0	18.2	10.5	9.9	21.6	10.0
3	226.5	7.9	6.3	7.3	6.1	2.6	2.6
4	219.8	5.2	5.8	9.4	5.0	2.0	2.4
5	151.3	3.5	3.5	6.9	4.9	1.4	1.4
6	127.7	2.7	0.0	4.4	2.1	2.1	0.7
B3							
1	1397.0	55.2	77.7	43.7	22.3	22.3	16.7
2	1411.0	43.6	62.3	33.8	18.8	5.0	11.0
3	1086.0	31.8	52.2	43.0	23.2	6.5	5.6
4	603.8	16.0	49.6	49.3	13.0	2.6	2.7
5	675.6	14.8	69.0	22.9	2.9	0.0	0.0
6	655.1	33.7	48.2	37.7	5.3	0.0	0.0
B4							
1	1476.0	37.1	76.6	26.8	0.8	10.3	1.6
2	1900.0	105.4	66.5	31.0	2.7	22.5	3.8
3	2811.0	175.5	97.9	66.3	4.9	32.2	7.7
4	1776.0	77.2	62.1	65.5	1.6	8.5	5.5
5	1244.0	61.1	41.7	66.4	0.0	2.1	3.6
	1213.0	22.5	25.5	28.6	1.6	0.0	3.3
B5							
1	2638.0	110.9	47.0	293.3	6.1	0.2	
2	2940.0	170.4	116.3	633.7	10.4	21.0	
3	1700.0	76.5	59.5	202.2	5.5	9.3	
4	1362.0	64.9	309.8	57.1	5.7	5.8	
5	923.7	71.4	9.0	0.0	5.0	0.0	
6	1254.0	196.0	10.8	21.7	5.6	3.1	
B6							
1	2084.0	140.8	433.4	103.7	9.3		
2	2433.0	152.6	310.8	100.4	20.7		
3	1358.0	79.5	348.0	114.1	28.0		
4	1689.0	116.6	431.3	97.5	25.9		
5	707.1	188.2	285.8	83.8	25.5		
6	758.6	44.5	265.5	85.0	16.3		
B7							
1	3476.0	86.7	77.5	0.0	5.6		
2	1776.0	35.8	64.5	3.5	4.9		
3	2054.0	34.5	22.9	6.9	2.1		
4	1507.4	8.8	84.9	90.2	6.8		
5	756.4	19.1	22.0	3.8	0.0		
6	1150.0	0.0	17.5	9.2	3.7		
B8							
1	1857.0	38.2	41.3	15.0	4.0	0.0	
2	2237.0	44.5	54.9	38.2	7.0	20.2	
3	1875.0	30.3	147.2	20.0	2.0	15.1	
4	1207.0	13.7	40.3	22.5	2.7	11.7	
5	967.5	1.5	30.7	5.0	0.0	0.0	
6	1650.0	31.0	50.3	15.0	2.7	0.0	

Table 4. Spherical moss bags: the percentage data for the Spearman's rank correlation coefficients between the concentrations of all metals over 47 sites in each batch; no coefficients were significantly negative.

Batch	Total no. coefficients	% coefficients significant	% at $p < 0.05$	% at $p < 0.005$
1	21	67	23	33
2	21	81	24	57
3	21	47	14	33
4	21	71	14	57
5	15	93	20	73
6	10	40	30	10
7	10	30	30	0
8	15	60	40	20

moss were used during the survey, and hence the unexposed SMB of the eight batches may have differed slightly in the replicability of their metal concentrations, pH values, and cation exchange capacities.

Comparative financial aspects of the study further underline the usefulness of SMB as a technique for low technology sampling. A 27-month study, using high technology samplers (pumps and filtering equipment) to monitor the concentrations of metals at four sites in Armadale, was undertaken shortly before the present study (*24*). Its outcome was unsatisfactory in two important ways: it failed to demonstrate the area of high pollution in the north of the town, because no sampler was placed there—probably as a result of financial reasons and the lack of suitable electric power points—and the difficulty in interpreting the findings, because results had been generated at only four sites, prevented the clear demonstration of a pollution gradient around the foundry (*3*). The cost of that survey is not known, but must have been high. By contrast, the materials for the present investigation using SMB, which allowed air pollution to be monitored at 47 sites within the town during 17 months, and which greatly facilitated the interpretation of epidemiological studies (*25*), cost less than £300, a fraction of what would have been incurred by a similar survey using high technology samplers.

Table 5. Spherical moss bags: the number of metals with which a given metal was significantly correlated in each batch.

Metal	Batch 1	Batch 2	Batch 3	Batch 4	Batch 5	Batch 6	Batch 7	Batch 8
Fe	5	6	4	6	5	2	2	4
Mn	5	5	4	5	4	3	2	5
Zn	5	4	2	5	5	2	2	3
Pb	4	3	0	4	5	0	0	1
Cu	5	5	4	2	5	1	0	2
Cr	4	5	2	4	4	—	—	3
Ni	0	6	4	4	—	—	—	—

Table 6. Spherical moss bags: the Spearman's rank correlation coefficients (r^2), and their significance values (p), between the mean concentrations of all metals over all batches in the 47 sites.

		Cr	Ni	Zn	Pb	Cu	Fe
Ni	r^2	0.54					
	p	0.001					
Zn	r^2	0.27	0.28				
	p	0.031	0.029				
Pb	r^2	0.65	0.38	0.34			
	p	0.001	0.004	0.010			
Cu	r^2	0.59	0.51	0.19	0.45		
	p	0.001	0.001	0.099	0.001		
Fe	r^2	0.60	0.65	0.54	0.64	0.52	
	p	0.001	0.001	0.001	0.001	0.001	
Mn	r^2	0.41	0.56	0.44	0.56	0.34	0.80
	p	0.002	0.001	0.001	0.001	0.010	0.001

Table 7. Spherical moss bags: the number of batches with which a given batch was significantly correlated for each metal.

Metal	Batch 1	Batch 2	Batch 3	Batch 4	Batch 5	Batch 6	Batch 7	Batch 8	Total no. of batches
Fe	7	5	6	5	6	7	5	7	8
Mn	7	4	5	3	5	5	1	6	8
Zn	3	2	1	5	3	0	3	3	8
Pb	3	0	3	2	2	3	0	1	8
Cu	0	0	2	1	0	0	0	1	8
Cr	1	3	1	2	0	—	—	1	6
Ni	0	1	1	0	—	—	—	—	4

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REFERENCES

1. Lloyd, O. L. Respiratory-cancer clustering associated with localized industrial air pollution. Lancet (i): 318–320 (1978).
2. Lloyd, O. L., Sclare, G., Lloyd, M. M., and Yule, F. A. Respiratory cancer in a Scottish community: some pathological, occupational and general environmental considerations. In: Cancer Campaign 6, Cancer Epidemiology (E. Grundmann, J. Clemmesen, and C. S. Muir, Eds.), Gustav Fischer Verlag, Stuttgart, 1982, pp. 103–114.
3. Lloyd, O. L., Barclay, R., Lloyd, M. M., and Armadale Group Practice. Lung cancer and other health problems in a Scottish industrial town: a review. Ambio 14:322–329 (1985).
4. Le Blanc, F., Robitaille, G., and Rao, D. Biological response of lichens and bryophytes to environmental pollution in the Murdochville coppermine area, Quebec. Hattori Bot. Lab. 38: 405–433 (1974).
5. Nieboer, E., and Richardson, D. H. S. Lichens as monitors of atmospheric deposition. In: Atmospheric Pollutants in Natural Waters (S. Eisenreich, Ed.), Ann Arbor Science, Ann Arbor, MI, 1981.
6. Margot, A., and Romain, M. Heavy metals in terrestrial cryptograms. A bibliographic synthesis. Mem. Soc. Roy. Bot. Belg. 7: 25–47 (1976).

7. Richardson, D. and Niebor, E. Nickel in lichens, bryophytes, fungi and algae. In: Nickel in the Environment (J. Nriagu, Ed.). John Wiley, New York, 1980.
8. Martin, H., and Coughtrey, P. Biological Monitoring of Heavy Metal Pollution. Applied Science, London, 1982.
9. Gailey, F. A. Y., and Lloyd, O. L. The use of *Lecanora conizaeoides* as a monitor of the distribution of atmospheric pollution by metals. Ecol. Disease 2: 215–224 (1983).
10. Yule, F. A., and Lloyd, O. L. The metal content of an indigenous moss in Armadale, Central Scotland. Water, Air Soil Pollut. 21: 261–270 (1984).
11. Gailey, F. A. Y., and Lloyd, O. L. Methodological investigations into low technology monitoring of atmospheric metal pollution. Part III. The consistency of the metal concentrations. Environ. Pollut., B12: 85–109 (1986).
12. Goodman, G., and Roberts, T. Plants and soils as indicators of metals in the air. Nature 231: 287–292 (1971).
13. Clymo, R. Ion exchange in *Sphagnum* and its relation to bog ecology. Ann. Botany 27: 309–324 (1963).
14. Makinen, A. Moss and peat-bogs in air pollution monitoring. Suo 28: 79–88 (1975).
15. Goodman, G., Smith, S., Parry, A., and Inskip, M. The use of moss bags as deposition gauges for airborne metals. Proceedings of the 41st Annual Conference of the National Society for Clean Air. Cardiff, October 14–18, 1974.
16. Muskett, C. A survey of airborne heavy metals in a city environment using biological indicators. Environ. Health 267–269 (November 1976).
17. Swaine, D., Goodbeer, W., and Morgan, N. Use of moss to measure the accession of trace elements to an area around a power plant. Proceedings International Conference on Heavy Metals in the Environment, Heidelberg, September 1983, II, pp. 1053–1056.
18. Gailey, F. A. Y., and Lloyd, O. L. Methodological investigations into low technology monitoring of atmospheric metal pollution. Part I. The effects of sampler size on metal concentrations. Environ. Pollut., B12: 41–59 (1986).
19. Yule, F. A., and Lloyd, O. L. Low technology monitoring of air pollution by metals in relation to the geography of lung cancer. In: Nickel in the Human Environment. (F. Sunderman, Ed.), IARC Scientific Publication No. 53, International Agency for Research on Cancer, Lyon, 1984, pp. 253–264.
20. Gailey, F. A. Y., and Lloyd, O. L. Grass and surface soils as monitors of atmospheric metal pollution in central Scotland. Water, Air Soil Pollut. 24: 1–18 (1985).
21. Gailey, F. A. Y., and Lloyd, O. L. A wind tunnel study of the flow of air pollution in Armadale, Central Scotland. Ecol. Disease 2: 419–431 (1984).
22. Schueneman, J., High, M., and Bye, W. Air pollution aspects of the iron and steel industry. Environmental Health Series, Public Health Series, 999-AP-1 U.S. Department of Health, Education and Welfare, Cincinnati, Ohio, 1963.
23. Puckett, K., Neiboer, E., Borzynski, M., and Richardson, D. The uptake of metal ions by lichens: a modified ion-exchange process. New Phytologist 72: 329–342 (1973).
24. Her Majesty's Industrial Pollution Inspectorate for Scotland. Multi-element survey in West Lothian; report on the results of the first twenty seven months. H.M.I.P.I., Edinburgh, 1981.
25. Lloyd, O. L., Williams, F., and Gailey, F. A. Y. Is the Armadale epidemic over? Air pollution and mortality from lung cancer and other diseases, 1961-1982. Brit. J. Ind. Med. 42: 815–823 (1985).